

Radiative heat transfer between dielectric bodies

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The recent development of a scanning thermal microscope (SThM) has led to measurements of radiative heat transfer between a heated sensor and a cooled sample down to the nanometer range. This allows for comparison of the known theoretical description of radiative heat transfer, which is based on fluctuating electrodynamics, with experiment. The theory itself is a macroscopic theory, which can be expected to break down at distances much smaller than 10^{-8} m. Against this background it seems to be reasonable to revisit the known macroscopic theory of fluctuating electrodynamics and of radiative heat transfer.

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The development of a new kind of scanning thermal microscope (SThM) by the experimental Energy and Semiconductor Research group in Oldenburg has made possible measurements of radiative heat transfer down to the nanometer range. A heated sensor tip incorporating a thermo-couple with a tip radius of 60 nm scans a cooled sample surface, registering the thermo-voltage in the sensor. In order to assure that the results are not influenced by convection or adsorbates on the sample surface, the SThM works under ultra high vacuum conditions. In the future, scanning thermal microscopy might become a technique supplementing the well-known scanning tunneling and atomic force microscopy. On the other hand, the measurements with the SThM allow for comparison of the existing theory of radiative heat transfer with the experiment. This shall be motivation enough for revisiting the theory of radiative heat transfer.

INTRODUCTION

The radiative heat transfer between macroscopic dielectric bodies is a well-understood physical phenomenon and the energy transported per unit time and unit area can be expressed by the Kirchhoff-Planck law [1] in the stationary case, i.e., when the bodies are in local thermal equilibrium. But this is only true for distances d which are large compared with the thermal wave length λ_{th} given by Wien's law, i.e., for $d \gg \lambda_{th}$. For distances $d \ll \lambda_{th}$ the radiative heat transfer increases strongly due to so-called evanescent modes.

For convenience we consider two semi-infinite bodies with a vacuum gap of distance d between them (see Fig. 1). This system is invariant under rotations and we chose the z -axis as the axis of rotation. We further assume that the first body at $z < 0$ has a temperature $T_1 \neq 0$, whereas the second semi-infinite body at $z > d$ has zero temperature, i.e., $T_2 = 0$. Due to quantum and vacuum fluctuations of the atoms and electrons in the first body there will be a fluctuating electromagnetic field inside that body. Now, the boundary conditions of macroscopic electrodynamics require that the transver-

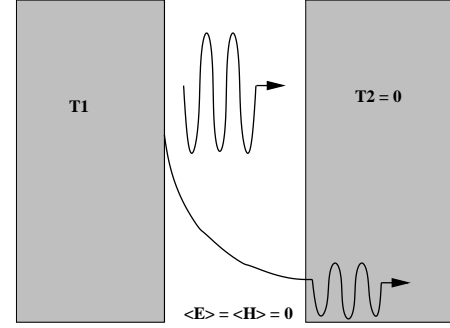


FIG. 1: Sketch of the propagating and evanescent modes contributing to the radiative heat transfer between two semi-infinite bodies at different temperatures.

sal components of the electric and the magnetic field are continuous at the interface $z = 0$. Therefore, there will also be a fluctuating electromagnetic field outside the semi-infinite body produced by fluctuating internal sources. This field can be divided into a propagating and an evanescent part. This becomes clear if we consider a plane wave solution $\exp(i(k_z z - \omega t))$ in the vacuum gap propagating into the positive z -direction, with the modulus of the wave vector given by

$$k_z = \sqrt{\frac{\omega^2}{c^2} - k_{\perp}^2} = \begin{cases} \text{real} & , k_{\perp} < \frac{\omega}{c}, \\ \text{complex} & , k_{\perp} > \frac{\omega}{c}. \end{cases} \quad (1)$$

Obviously, the modulus of the wave vector k_z is purely real if the modulus of the transversal wave vector k_{\perp} is smaller than the modulus of the vacuum wave vector, $k_0 = \omega c^{-1}$. In this case we will have an oscillatory solution, i.e., propagating waves. On the other hand, k_z is purely imaginary if $k_{\perp} > k_0$. In that case the plane wave is exponentially damped in z -direction, i.e., the solution is a so-called evanescent wave.

The characteristic wave length of the fluctuating field in the vacuum gap, generated in the semi-infinite body at $z < 0$, is the thermal wave length λ_{th} . It follows that for distances $d \gg \lambda$, i.e., in the far-field region, the evanescent field can not reach into the second body and

therefore only propagating modes can contribute to the radiative energy transfer. This energy transfer — as mentioned above — can then be described with the Kirchhoff-Planck law and is smaller than the energy transfer between two black bodies, which is given by the Stefan-Boltzmann law [1]

$$S_{\text{BB}} = \sigma(T_1^4 - T_2^4) \quad (2)$$

with $\sigma = 5.67 \cdot 10^{-8} \text{Wm}^{-2}\text{K}^{-4}$. On the other hand, for distances d between the two bodies smaller than λ_{th} , i.e., in the near-field region, the evanescent waves generated in the first body can reach into the second body and propagate there. This can be seen as some kind of photon tunneling through a vacuum barrier (see Fig. 1). Hence, in the near-field the propagating and evanescent fluctuating fields both will contribute to the radiative energy transfer. In that case, the energy transfer can be several orders of magnitude larger than the black body value (2), as we will see later.

THEORETICAL DESCRIPTION

Fluctuating fields

The theoretical treatment of radiative heat transfer is usually based on Rytov's fluctuating electrodynamics [2]. Within this framework the macroscopic Maxwell equations are augmented by fluctuating source currents \mathbf{j} , describing the fluctuating sources of the electric and magnetic field \mathbf{E} and \mathbf{H} inside the dielectric dissipative body. The individual frequency components of these source currents $\mathbf{j}(\mathbf{r}, \omega)$ are considered as stochastic Gaussian variables. Within this treatment the Maxwell equations become so-called stochastic Maxwell equations

$$\begin{aligned} \nabla \times \mathbf{E}(\mathbf{r}, \omega) &= i\omega\mu_0\mathbf{H}(\mathbf{r}, \omega), \\ \nabla \times \mathbf{H}(\mathbf{r}, \omega) &= \mathbf{j}(\mathbf{r}, \omega) - i\omega\epsilon(\omega)\mathbf{E}(\mathbf{r}, \omega) \end{aligned} \quad (3)$$

with the permeability of the vacuum μ_0 and the permittivity of the body $\epsilon(\omega)$. By writing the stochastic Maxwell equations in the form of eq. (3) the dielectric body containing the source currents is already assumed to be non-magnetic, homogeneous, isotropic and local.

The electric and magnetic fields described by the stochastic Maxwell equations are classical stochastic processes and can formally be expressed as

$$\begin{aligned} \mathbf{E}(\mathbf{r}, \omega) &= i\omega\mu_0 \int d^3r' \mathbf{G}^E(\mathbf{r}, \mathbf{r}') \mathbf{j}(\mathbf{r}', \omega), \\ \mathbf{H}(\mathbf{r}, \omega) &= i\omega\mu_0 \int d^3r' \mathbf{G}^H(\mathbf{r}, \mathbf{r}') \mathbf{j}(\mathbf{r}', \omega) \end{aligned} \quad (4)$$

where the integral has to be taken over the source region, i.e., over the volume of the dielectric body containing the source currents. The tensors \mathbf{G}^E and \mathbf{G}^H are the dyadic electric and the magnetic Green's function. The electric

dyadic Green's function is a solution of the Helmholtz wave equation

$$(\nabla \times \nabla \times - k^2) \mathbf{G}^E(\mathbf{r}, \mathbf{r}') = \mathbb{1} \delta(\mathbf{r} - \mathbf{r}') \quad (5)$$

with the wave vector \mathbf{k} and the unit matrix $\mathbb{1}$. By definition, the electric dyadic Green's function satisfies the boundary conditions of the electric field. Even though a similar statement holds for the magnetic Green's function, it is more convenient to construct the dyadic magnetic Green's from the electric Green's function by means of the relation

$$\mathbf{G}^H(\mathbf{r}, \mathbf{r}') = -\frac{i}{\omega\mu_0} \nabla \times \mathbf{G}^E(\mathbf{r}, \mathbf{r}'), \quad (6)$$

which is a direct consequence of Maxwell's equations.

The reformulation of the electric and magnetic field \mathbf{E} and \mathbf{H} as an integral over the source currents in eqs. (4) makes clear that the fluctuating properties of the fields are directly determined by the fluctuational properties of the source currents. In fact, if $\langle \mathbf{j}(\mathbf{r}, \omega) \rangle$ and $\langle \mathbf{j}(\mathbf{r}, \omega) \mathbf{j}^*(\mathbf{r}', \omega') \rangle$ are known, where the angular brackets symbolise the ensemble average, the corresponding averages and correlation functions of the fields can be evaluated. Therefore, it remains to determine the stochastic properties of the source currents.

From the definition of the source currents as stochastic Gaussian variables describing the thermal and quantum fluctuations inside a dielectric dissipative body, it should be clear that the ensemble average of the source currents vanishes. It follows with (4) that

$$\langle \mathbf{E}(\mathbf{r}, \omega) \rangle = \mathbf{0} \quad \text{and} \quad \langle \mathbf{H}(\mathbf{r}, \omega) \rangle = \mathbf{0}. \quad (7)$$

However, the correlations and therewith the fluctuations of the source currents are determined by the properties of the dielectric dissipative body. This connection between dissipation and fluctuation can be expressed in a general way by means of the fluctuation-dissipation theorem [1], which can be applied to our problem [2]. For a homogeneous, isotropic and local dielectric dissipative body this yields

$$\begin{aligned} \langle j_\alpha(\mathbf{r}, \omega) j_\beta^*(\mathbf{r}', \omega') \rangle \\ = 4\pi\omega E(\omega, T) \epsilon''(\omega) \delta_{\alpha\beta} \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega') \end{aligned} \quad (8)$$

for the components of the source currents, where we have written $\epsilon = \epsilon' + i\epsilon''$, and $E(\omega, T)$ is the Einstein function

$$E(\omega, T) = \frac{\hbar\omega}{2} + \frac{\hbar\omega}{e^{\hbar\omega\beta} - 1} \quad (9)$$

with the usual inverse temperature $\beta = 1/k_B T$ of the body. The appearance of the Einstein function is implied by the fluctuation-dissipation theorem, which indeed is a quantum mechanical operator relation, but the fluctuating fields in the framework of fluctuating electrodynamics

are classical. This is a somewhat weak point in the theory, but it was recently shown [3] that a rigorous quantum electrodynamical treatment for dissipative media leads to the same correlation functions for the fields as fluctuating electrodynamics. Hence, it seems to be justified to work within the framework of fluctuating electrodynamics, which should be classified as a semi-classical theory.

Now, it is possible to evaluate the correlation functions of the fluctuating fields. Using eqs. (4) together with the fluctuation-dissipation theorem in eq. (8) yields

$$\begin{aligned} \langle E_\alpha(\mathbf{r}, t) H_\beta(\mathbf{r}, t) \rangle &= \int_0^\infty d\omega E(\omega, T) \frac{\omega \epsilon''(\omega)}{\pi} (\mu_0^2 \omega^2) \\ &\times \int d^3 r' \left(\mathbf{G}^E \mathbf{G}^{H\dagger} \right)_{\alpha\beta} + \text{c.c.} \end{aligned} \quad (10)$$

where the \dagger -symbol denotes the adjoint dyadic and c.c. is an abbreviation for the complex conjugated term. Similar equations can be derived for $\langle E_\alpha(\mathbf{r}, t) E_\beta(\mathbf{r}, t) \rangle$ and $\langle H_\alpha(\mathbf{r}, t) H_\beta(\mathbf{r}, t) \rangle$, respectively. The ensemble averages of the physical quantities — the Poynting vector, the energy density and the stress tensor — of a dielectric dissipative body can be evaluated with the help of these field correlation functions, if the material properties, i.e., the permittivity $\epsilon(\omega)$, and the temperature of the body are known. Furthermore, it is necessary to calculate the dyadic Green's functions for the given electrodynamical problem, in which the geometry of the problem is incorporated.

Radiative heat transfer between two slabs

In principle, we are now able to calculate the radiative heat transfer between dielectric bodies of arbitrary shape and volume as long these bodies are homogeneous, isotropic, and local dissipative dielectrics, and can be described macroscopically. Unfortunately, the calculation of the dyadic Green's function for the most geometries is rather complicated. Therefore, we will consider only the radiative heat transfer between two bodies of the simplest possible geometry, i.e., two semi-infinite dielectric bodies separated by a vacuum gap of width d (see Fig. 1).

The solution to this problem is well-known: the first derivation in the framework of Rytov's fluctuating electrodynamics was given by Polder and van Hove [4]. Levin *et al.* [5] solved the problem with the surface impedance method of Leontovich, and Loomis and Maris [6] reinvestigated the problem some years ago. The radiative heat transfer between the two slabs can be stated as [4]

$$\begin{aligned} \langle S \rangle &= \frac{1}{4\pi^2} \int_0^\infty d\omega [E(\omega, T_1) - E(\omega, T_2)] \\ &\times \int_0^\infty dk_\perp k_\perp \{T_\parallel^{12} + T_\perp^{12}\} \end{aligned} \quad (11)$$

where T_\parallel^{12} and T_\perp^{12} are the transmission coefficients from the first body at $z < 0$ to the second body at $z > d$ for the TM- and TE-modes, respectively. These transmission coefficients can be expressed by means of the usual Fresnel coefficients [7], which gives for the propagating modes, i.e., for $k_\perp < k_0$,

$$(T_\parallel^{12})^{\text{prop}} = \frac{(1 - |r_\parallel^1|^2)(1 - |r_\parallel^2|^2)}{|1 - r_\parallel^1 r_\parallel^2 e^{2ik_z d}|^2}, \quad (12)$$

and for the evanescent modes, i.e., for $k_\perp > k_0$,

$$(T_\parallel^{12})^{\text{ev}} = \frac{4\text{Im}(r_\parallel^1)\text{Im}(r_\parallel^2)e^{-2\gamma d}}{|1 - r_\parallel^1 r_\parallel^2 e^{-2\gamma d}|^2}, \quad (13)$$

where the transmission coefficients for the TE-modes can be deduced from these relations by the substitution $\parallel \rightarrow \perp$.

The mean energy transfer per unit time and unit area between the two semi-infinite bodies at different temperatures in eq. (11) consists of two parts. The temperature enters only into the first part, which is given by the difference of the Einstein functions evaluated at the corresponding temperatures. From this difference it becomes clear that the vacuum term in the Einstein functions vanishes; there will be no vacuum contribution to the energy transfer. The second part is the integral over the lateral wave vector k_\perp which counts the modes contributing to the energy transfer. It can be shown that the above transmission coefficients are always smaller than 1. Moreover, it is possible to retrieve the energy transfer between two black bodies, which by definition have zero Fresnel coefficients. It follows that the transmission coefficient for the evanescent modes (13) becomes zero and the transmission coefficient for the propagation modes (12) becomes 1. Inserting these transmission coefficients in eq. (11) yields the Stefan-Boltzmann law stated in eq. (2).

From eq. (11) together with the transmission coefficients it is possible to study the radiative heat transfer between the two semi-infinite bodies in detail. At the moment, we are only interested in the near-field limit of eq. (11), i.e., in distances $k_0 d \ll 1$. A Taylor expansion [5] shows that in the near-field

$$\langle S_\parallel \rangle \propto \frac{1}{d^2} \quad \text{and} \quad \langle S_\perp \rangle = \text{const.} \quad (14)$$

Therefore, the TM-mode part of the energy transfer dominates in the near-field region, $\langle S_\parallel \rangle \gg \langle S_\perp \rangle$, but it is not clear a priori at what distance this domination begins. In fact, we will see by numerical calculations that the distance where the TM-mode contribution becomes dominant depends on the material properties of the two slabs. Furthermore, from the near-field limit in eq. (14) two questions arise: i) Is a divergent radiative energy transfer physically reasonable? ii) Is it possible to measure this power law? Or better, at what distance does the domination of the TM-mode part begin?

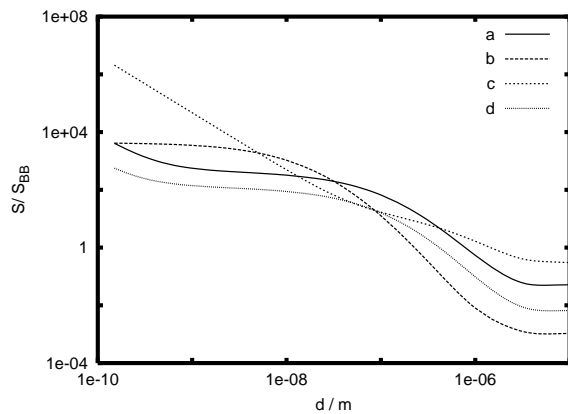


FIG. 2: Numerical results of eq. (11) for different Drude materials at different temperatures normalised to the black body value S_{BB} given by the Stefan-Boltzmann law in eq. (2).

The first question was discussed controversially [8–10]. But it should be clear that the description based on fluctuating electrodynamics is still a macroscopic one, which means that at distances smaller than 10^{-8}m this theory is not valid and therefore does not lead to physically reasonable results. In fact, the source currents in the fluctuation-dissipation theorem in eq. (8) are delta-correlated with respect to $\mathbf{r} - \mathbf{r}'$, but this can not be true at distances where the microscopic properties of the materials make themselves felt. This delta-correlation of source currents seems to be the source of the divergent heat transfer at small distances, but in order to give a satisfactory answer to the question stated above a theory is needed that takes the finite correlations of source currents into account.

With this drawback of the macroscopic theory and the second question in mind we will now discuss some numerical results of the heat transfer (11) between two Drude materials [11]. For convenience we set the temperature of the first body to $T_1 = 300\text{K}$ and the temperature of the second body to $T_2 = 0\text{K}$. Moreover, we use the same Drude permittivity for both bodies, i.e., $\epsilon_1 = \epsilon_2$.

The numerical results for different plasma frequencies ω_p and relaxation times τ are given in Fig. 2. For a , b , and d we used relatively high relaxation times and high plasma frequencies, so that a , b , and d are plots for good conductors. On the other hand, for c we used a relatively small relaxation time and small plasma frequency, which means that c is a plot for a bad conductor.

Obviously the divergency discussed before, which is associated with the TM-mode part of the radiative heat transfer $\langle S_{\parallel} \rangle$, can only be seen for c , i.e., for the bad conductor. There also is a divergency for good conductors, but it appears only for distances much smaller than 10^{-9}m . Otherwise, in the region between 10^{-9}m and 10^{-7}m the radiative heat transfer for the good conductors a , b , and d becomes constant. As discussed before, such a

behaviour can be attributed to the TE-mode part of the radiative heat transfer, $\langle S_{\perp} \rangle$. As an answer to the second question, it follows that for bad conductors the radiative heat transfer in the region between 10^{-9}m and 10^{-7}m , which is accessible to measurements, is dominated by the TM-mode part of the radiative heat transfer, whereas for good conductors the TE-mode part dominates.

SUMMARY AND OUTLOOK

The radiative heat transfer between macroscopic dielectric bodies can be described by Rytov's theory of fluctuating electrodynamics. In the near-field the evanescent modes give the main contribution to the radiative heat transfer. As the numerical results indicate, for good conductors the TE-mode part dominates the heat transfer for experimentally accessible distances. In contrast, for bad conductors the TM-mode part dominates in that region and leads to a divergent radiative heat transfer. This unsatisfactory feature of the theory can be traced back to the delta correlation in the correlation function of the source currents. There is a need for a theory which takes a finite correlation length of the source currents into account, and which should lead to a finite heat transfer at all distances.

Experiments now being prepared in Oldenburg should answer some questions and thus provide a basis for such a theory. Some of these questions are: At what distance does the macroscopic theory fail? What is the finite value of $\langle S \rangle$ for $d \rightarrow 0$? Is there an appropriate theory describing the sensor-sample geometry?

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